

# A numerical study of two-photon ionization of helium using the Pyprop framework

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Few-photon induced breakup of helium is studied using a newly developed *ab initio* numerical framework for solving the six-dimensional time-dependent Schrödinger equation. We present details of the method and calculate (generalized) cross sections for the process of two-photon nonsequential (direct) double ionization at photon energies ranging from 39.4 to 54.4 eV, a process that has been very much debated in recent years and is not yet fully understood. In particular, we have studied the convergence property of the total cross section in the vicinity of the upper threshold ( $\sim 54.4$  eV), versus the pulse duration of the applied laser field. We find that the cross section exhibits an increasing trend near the threshold, as has also been observed by others, and show that this rise cannot solely be attributed to an unintended inclusion of the sequential two-photon double ionization process, caused by the bandwidth of the applied field.

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## I. INTRODUCTION

The study of how light interacts with matter has occupied physicists for a long time. Of particular fundamental interest is single and multiple ionization of atoms and molecules by photon impact, with subsequent ejection of one or a multiple number of electrons. In this respect, single-photon multiple ionization is special, as exchange of energy between the involved electrons is a prerequisite for the process to take place. The investigation of such correlated dynamical processes poses many unique challenges to experiment and theory. A prime example of this is the one-photon double ionization of helium, which has been the subject of intense study since the pioneering work of Byron and Joachain [1]. As a matter of fact, it was only recently that a complete agreement between theoretical calculations and accurate measurements with synchrotron radiation was established, for the value of the (generalized) cross section for the direct (nonsequential) double ionization process [2–5].

The problem of two-photon double ionization of two-electron atomic systems presents additional difficulties. First, the separation between sequential and nonsequential double ionization often becomes a nontrivial problem [6], in situations where both processes are energetically accessible [7]. Here, 'sequential' ionization usually refers to the process where the electrons are emitted one after the other by subsequent absorption of a photon each, and where the second electron has time to relax to a stationary ionic state before it is emitted. Thus, energy exchange between the two electrons is not strictly required. In contrast, 'nonsequential' double ionization depends upon exchange of energy between the outgoing electrons, and as such it represents a clear departure from an independent-particle picture. As mentioned above, after more than 40 years of investigation, the case of double electron ionization by single photon absorption is now considered

to be a well understood process [8], but the related problem of two-photon direct (nonsequential) double ionization in helium, at photon energies ranging from 39.4 to 54.4 eV, is still being debated [5, 9–22]. In particular, the separation of two-photon single ionization, where the remaining electron is left in an (excited) ionic state, from two-photon double ionization, has turned out to be a subtle theoretical problem, as the role of electron correlations in the final states is not yet fully understood [5]. Moreover, moving beyond the single-photon ionization regime is extremely challenging from the experimental point of view, due to the weakness of the signals. Although the total cross section for the two-photon nonsequential double ionization of helium has been the subject of experiments, employing state of the art high-order harmonic [23, 24] and free-electron laser (FEL) radiation [25], the experimental results remain inconclusive [26].

In this paper, we revisit the problem of two-photon nonsequential double ionization in helium. We present details on a recently developed B-spline based numerical framework for solving the two-electron time-dependent Schrödinger equation. The method is built on the more general Pyprop framework [27] and was recently used to study the role of electron correlations in the stabilization of helium in intense extreme-ultraviolet (XUV) laser fields [28]. The two-electron module we have implemented is designed to utilize massively parallel supercomputers to perform accurate large-scale time-dependent simulations, and we here use it to make a contribution to the ongoing discussion on two-photon double ionization.

For the total cross section we obtain values that are in close agreement with some recently reported results [18, 21]. In contrast, our results are in clear disagreement with results from calculations where correlation effects in the final scattering states have been included to some extent, using different approximative methods. In our approach, which is similar to the method adapted by Feist *et al.* [18], and others, this problem is circumvented by letting the ionized wave packet propagate for some time after the end of the laser pulse, so that most of the wave packet eventually reaches the asymptotic (Coulomb) region [29]. The wave function is then analyzed by means of projections onto a set of uncorrelated eigen-

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states, i.e., Slater determinants constructed from one-electron orbitals.

With our present method we are able to consider relatively long pulses (up to about 10 fs in total pulse duration). This puts us in a position to study the convergence property of the total cross section in the vicinity of the upper threshold ( $\sim 54.4$  eV), as a function of pulse length. It has been reported that the cross section exhibits an apparent sharp rise near the threshold [14, 16, 18, 21, 30]. This rise stands somewhat in contrast to the results obtained in lowest (nonvanishing) order perturbation theory (LOPT) [6]. Our calculations with longer pulses indicate that the cross section indeed appears to grow as it approaches the threshold (up to 51.7 eV), but we cannot rule out the possibility that the cross section reaches a maximum at some point in the immediate neighborhood of (or on) the threshold. Based on our results we are tempted to conclude that the increase of the cross section around 54.4 eV is not solely due to an unintended inclusion of the sequential process [6, 30]. If this is correct, the very interesting question remains: what is the underlying physical process causing the unexpected behavior?

The rest of this paper is organized into two main sections. The first outlines some theoretical background, and then goes on to discuss the various aspects of the numerical method, ending with a discussion of convergence issues. In the final section, we present our calculations of the total cross section, and pay particular attention to its behavior near the sequential threshold.

Atomic units ( $\hbar$ ,  $m_e$  and  $e$  replaced by 1) are used in the following exposition, except where otherwise noted.

## II. THEORY AND NUMERICAL APPROACH

B-splines [31] have long been popular in atomic and molecular physics computations, due to their ability to accurately represent atomic eigenstates (see [32] and references therein). For time-dependent calculations, integration of the Schrödinger equation directly in the B-spline representation is very efficient for one-electron systems, due to the sparse and structured matrices that arise [33]. For two-electron systems, however, the matrix structure becomes more complicated, and the matrix sizes are also much larger. Therefore, in time-dependent approaches for two-electron systems using B-spline discretization, one-electron orbitals or atomic eigenstates have been used to construct a matrix representation of the field interactions. Both these approaches are useful and accurate, but do not easily scale to very large basis sizes, because the basis functions are global, resulting in dense matrices. Operations involving such matrices are difficult to parallelize efficiently, which eventually becomes necessary as the basis size is increased.

In this section, we present some details of a recently developed B-spline based numerical approach to the two-electron time-dependent Schrödinger equation, where the time integration is performed directly in the B-spline basis. A Python/C++ implementation of the method have been created, which uses the Pyprop framework [27].

### A. Theoretical background

We consider the two-electron time-dependent Schrödinger equation (TDSE),

$$i \frac{\partial}{\partial t} \Psi(\mathbf{r}_1, \mathbf{r}_2, t) = H\Psi(\mathbf{r}_1, \mathbf{r}_2, t). \quad (1)$$

Employing the semi-classical approximation, the light-atom interaction Hamiltonian can be cast into the form,

$$H = \left( \frac{\mathbf{p}_1^2}{2} - \frac{2}{r_1} + H_{f,1} \right) + \left( \frac{\mathbf{p}_2^2}{2} - \frac{2}{r_2} + H_{f,2} \right) + \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}, \quad (2)$$

where  $H_{f,i}$  represents the interaction with the external field. The laser-atom interaction is modeled in the dipole approximation using the velocity gauge formulation, which, when linearly polarized along the  $z$  axis, can be written as

$$H_{f,i} = A_z(t)p_{z_i}. \quad (3)$$

Here  $A_z$  is the vector potential defining the external field. The corresponding electric field is given by  $E_z = -\partial A_z / \partial t$ . For the temporal form of the laser interaction, a sine-squared carrier-envelope was chosen, i.e.,

$$A_z(t) = A_0 \sin^2\left(\frac{\pi t}{T}\right) \cos(\omega t), \quad (4)$$

where  $A_0 = E_0/\omega$ ,  $E_0$  being the electric field amplitude,  $\omega$  is the (central) laser frequency, and  $T$  defines the (total) pulse duration. We have also considered pulses with a Gaussian envelope,

$$A_z(t) = A_0 \exp\left[-2 \ln 2 \left(\frac{t-t_0}{\tau_0}\right)^2\right] \cos[\omega(t-t_0)], \quad (5)$$

where  $\tau_0$  is the full-width at half-maximum (FWHM) pulse duration, and  $T = 2t_0$  defines the (chosen) total pulse duration.

### B. Discretization

Turning the continuous TDSE into a set of coupled ordinary differential equations is achieved by representing the wave function, in spherical coordinates, as a product of radial B-splines and Coupled Spherical Harmonics,

$$\psi(\mathbf{r}_1, \mathbf{r}_2, t) = \sum_{i,j,k} c_{ijk}(t) \frac{B_i(r_1)}{r_1} \frac{B_j(r_2)}{r_2} \mathcal{Y}_{l_1, l_2}^{LM}(\Omega_1, \Omega_2). \quad (6)$$

Here,  $k = \{L, M, l_1, l_2\}$  is a combined index for the angular indices, and the Coupled Spherical Harmonics are given in

terms of Spherical Harmonics as [34]

$$\mathcal{Y}_{l_1 l_2}^{LM} = \sum_m \langle l_1 l_2 m M - m | LM \rangle Y_{l_1}^m(\Omega_1) Y_{l_2}^{M-m}(\Omega_2). \quad (7)$$

For the special case of  $z$ -polarization, the problem reduces effectively to five dimensions, as  $M$  is conserved during the laser-atom interaction. Since we are studying ionization from the ground state ( $M = 0$  manifold), only  $M = 0$  is included in the calculations. The B-spline basis functions depend upon several parameters, and determining the optimal values of these are not trivial (see Bachau *et al.* [32] for a discussion). Throughout this paper, we have used order 5 B-splines, and an exponential distribution of breakpoints near the origin, with linear spacing further out, providing accurate representation of both bound and continuum states. Zero boundary conditions are imposed by removing the first and last B-spline for each radial direction.

Except for the electron-electron interaction, all the terms in Eq. (2) are one-electron operators, and are straightforward to discretize. When calculating matrix elements of these, the radial integrals are performed with Gauss-Legendre quadrature [32], while the angular integrals are handled analytically. The electron-electron interaction is first expanded in a truncated multipole series,

$$\frac{1}{r_1 - r_2} \approx \sum_{l=0}^{l_{max}} \sum_{m=-l}^l \frac{4\pi}{2l+1} \frac{r_<^l}{r_>^{l+1}} Y_{l,m}^*(\Omega_1) Y_{l,m}(\Omega_2), \quad (8)$$

and each of the terms are handled in a similar manner to the one-electron operators. Finally, taking into account the non-orthogonality of the B-spline basis,  $\int dr B_i(r) B_j(r) = S_{ij}$ , the TDSE can be written in matrix form

$$iS \frac{\partial}{\partial t} \mathbf{c}(t) = \mathbf{H}(t) \mathbf{c}(t). \quad (9)$$

The total overlap matrix  $\mathbf{S}$  is a Kronecker product of one-electron overlap matrices for each angular momentum component,

$$\mathbf{S} = \mathbf{I}_k \otimes \mathbf{S}_1 \otimes \mathbf{S}_2. \quad (10)$$

In contrast to the one-electron case, the Hamiltonian matrix  $\mathbf{H}$  does not have a simple banded structure, although it is quite sparse. An overall banded structure remains, but is now interlaced with bands of zeros.

At present, we make use of multiprocessor systems by distributing the wavefunction across several processor in the angular rank, such that all the time-independent radial matrix elements are processor-local. This of course restricts the number of processors we may use to the total number of angular momentum elements. We are currently implementing the option of distributing one or both radial ranks, which would allow us to use more processors, and thus an even larger radial basis.

### C. Time integration

It is common for numerical integrations schemes to be based on the first-order exponential approximation to the propagator,

$$T(t_i, t_f) = \exp [-i\mathbf{S}^{-1} \mathbf{H} \Delta t] + \mathcal{O}(\Delta t^2), \quad (11)$$

which is quite accurate for reasonably small time steps  $\Delta t = t_f - t_i$ . The matrix exponential may be calculated efficiently by the popular Arnoldi or Lanczos methods [18, 35, 36]. However, instabilities prevent us from using this approach in the present case; instead, we use the implicit Cayley-Hamilton form of the propagation operator,

$$\left( \mathbf{S} + \frac{i\Delta t}{2} \mathbf{H} \right) \mathbf{c}(t + \Delta t) = \left( \mathbf{S} - \frac{i\Delta t}{2} \mathbf{H} \right) \mathbf{c}(t). \quad (12)$$

The above linear system of equations is typically too large to be solved by direct methods, but very sparse. It is therefore solved using the iterative Generalized Minimum Residual method (GMRES) [37, 38]. Similarly to the Arnoldi method, GMRES uses a Krylov subspace, constructed from successive multiplications of  $(\mathbf{S} + \frac{i\Delta t}{2} \mathbf{H})$  on  $\mathbf{c}(t)$  in each time step. A least-square problem is solved in the subspace spanned by these vectors, and a solution of the equation with a minimum residual is obtained. With this method, the error in the computed solutions (residual) is controlled by the size of the Krylov subspace, which can be increased automatically, thus always ensuring a high precision solution.

As is typical for discretized partial differential equations, and in particular those obtained with a B-spline basis, the system is quite stiff, and a good preconditioner is essential for the convergence of GMRES. Let  $\mathbf{A} = (\mathbf{S} + \frac{i\Delta t}{2} \mathbf{H})$  and  $\mathbf{b} = (\mathbf{S} - \frac{i\Delta t}{2} \mathbf{H})\mathbf{c}$ . The preconditioner  $\mathbf{M}$  is then constructed to make the linear system

$$\mathbf{M}^{-1} \mathbf{A} \mathbf{c} = \mathbf{M}^{-1} \mathbf{b} \quad (13)$$

easier to solve than the original system. This can be achieved by letting  $\mathbf{M}$  be an approximation of  $\mathbf{A}$ . The preconditioner used in this paper is a block type preconditioner, where each block consists of the complete radial Hamiltonian for a given coupled spherical harmonic ( $k$  is the angular index),

$$M_{(i,j,k),(i',j',k')} = \left( S_{(i,j,k),(i',j',k)} + \frac{i\Delta t}{2} H_{(i,j,k),(i',j',k)} \right) \delta_{k,k'}. \quad (14)$$

This block diagonal matrix is distributed across processors in such a manner that the elements in the wave function corresponding to one block are all local to one processor. When solving linear systems involving  $\mathbf{M}$ , each block can be solved separately and thus there is no communication between different processors. Furthermore, as the exact solution of  $\mathbf{M}$  is not required, we employ the incomplete LU (ILU) factorization [38],  $\mathbf{M} \approx \mathbf{LU}$ , as provided by the IFPACK toolkit available in the Trilinos [39] library. A similar preconditioner, using SuperLU [40] to solve  $\mathbf{M}$  exactly, was also tested, but found to be less efficient for the given system.

### D. Extracting physical information

Although excitation of the neutral atom is negligible for the intensities and frequencies we will consider in this paper, calculation of a subset of the eigenstates of the helium atom is nevertheless useful in many cases. The implicitly restarted Arnoldi method (IRAM) [41] may be used for this purpose. For reasons similar to those prompting the use of a preconditioner above, IRAM will converge slowly for interior eigenvalues. However, shifted inverse iterations can be used to accelerate the convergence for eigenvalues near a given shift  $\sigma$ ,

$$(\mathbf{H} - \sigma \mathbf{S})^{-1} \mathbf{c}_n = \frac{1}{E_n - \sigma} \mathbf{c}_n \mathbf{S}. \quad (15)$$

IRAM requires the multiplication of the operator matrix on a vector in order to operate. For inverse iterations this corresponds to solving the linear system in Eq. (15), for the matrix  $\mathbf{B} = (\mathbf{H} - \sigma \mathbf{S})$ . Note the similarity between  $\mathbf{A}$  and  $\mathbf{B}$ , the difference being only the scalars in front of  $\mathbf{H}$  and  $\mathbf{S}$ . The linear solver used for the propagation can therefore also be used to find eigenvalues with IRAM. Alternatively, a preconditioned Davidson method can be used. We found that when our basis grows sufficiently large, the Block-Davidson approach [42], as implemented in the Trilinos package Anasazi, performed favorably compared to the shift-invert Arnoldi method. In most cases, either of these methods can be used to rapidly obtain eigenpairs in the vicinity of any given shift value.

In order to extract double ionization probabilities from the wavefunction, we must project onto some set of states which span this space. The exact double continuum states are hard to find, as they require solving a scattering problem for the full two-particle system. An approximation using non-correlated states, obtained by solving a set of one-electron radial eigenvalue problems, is used instead,

$$\left( -\frac{1}{2} \frac{\partial^2}{\partial r^2} + \frac{l(l+1)}{2r^2} - \frac{Z}{r} \right) R_{n,l}^Z(r) = E_{n,l}^Z R_{n,l}^Z(r). \quad (16)$$

The double ionization continuum is represented by a product of  $\text{He}^+$  ( $Z = 2$ ) states, which, when expanded in the B-spline basis, we will denote  $\mathbf{b}_{n_1, n_2, k}$ . These states are not orthogonal to the bound states of the atomic system, and consequently the projection of the final wave function on the atomic bound states should be removed before the analysis is performed. Furthermore, the approximated double continuum states used here are not eigenstates of helium as the electron-electron correlation is ignored. The wave packet must therefore be propagated after the interaction until it reaches the Coulomb zone [29], where the electron-electron interaction is negligible.

To calculate the double ionization probability, we project the final wave function onto the non-correlated double continuum functions  $\mathbf{b}$ , and sum over all contributions,

$$P_{\text{double}} = \sum_{n_1, n_2, k} |\mathbf{b}_{n_1, n_2, k} \cdot \mathbf{c}(T)|^2. \quad (17)$$

Having found the double ionization probability, we may then

determine the total cross section for the nonsequential two-photon double ionization process [18, 43],

$$\sigma_{DI} = \left( \frac{\omega}{I_0} \right)^2 \frac{P_{\text{double}}}{T_{\text{eff}}}, \quad (18)$$

where  $I_0$  is the pulse intensity. The finite duration of the pulse is accounted for by  $T_{\text{eff}}$ , which for a two-photon process reads [18, 43]

$$T_{\text{eff}} = \int_{-\infty}^{\infty} \left[ \frac{I(t)}{I_0} \right]^2 dt. \quad (19)$$

For a sine-squared pulse envelope  $T_{\text{eff}} = 35T/128$ , while for a Gaussian envelope  $T_{\text{eff},g} = \tau_0/2\sqrt{\pi/2 \ln 2}$ .

### E. Accuracy and numerical convergence

The reliability of numerically calculated quantities must be checked carefully, and this is usually performed by varying the relevant numerical parameters and studying the resulting changes. It is possible, from certain physical considerations, to obtain a reasonable *a priori* estimate for the values of some parameters. In other cases, simple numerical calculations may be performed to get such estimates. For example, in the present case the number of photons absorbed in the system determines the maximum value of the total angular momentum quantum number  $L$  that must be included in the basis, and also the required radial box size, from an estimate of the energy of the ejected electrons. The remaining radial basis parameters mainly determine the quality of the ground state (see Table I), and the maximum photoelectron energy supported. By solving the related one-dimensional radial eigenvalue problem (Coulombic potential,  $Z = 2$ ), estimates for these parameters can be obtained. Specifically, estimating the density of states from  $1/\Delta E_n$ ,  $\Delta E_n$  being the energy separation between state number  $n$  and  $n+1$  in the discretized basis, and comparing with the known value, we may determine to what extent the density of states is correctly represented in the box, in the energy region of interest. Finally, by sampling the parameter space in the vicinity of the values thus estimated, a good indication as to whether the results are converged is obtained.

Time step size must also be considered, since our implicit time integrator incurs a local error of order  $\Delta t^3$ . Incidentally, this error is one order of magnitude smaller than that of the exponential propagator it approximates, but the constants are typically different, and may depend on all the other parameters except  $t$ . In any case, we have used a default time step  $\Delta t = 0.01$  a.u. Halving the time step to 0.005 a.u. produced changes in our calculated quantities of less than 0.1%.

Quantity	Calculated value (a.u.)	Reference value (a.u.)
He ( $1^1S$ )	-2.903 667	-2.903 724
He ( $2^1S$ )	-2.145 971	-2.145 974
He ( $1^3S$ )	-2.175 229	-2.175 229
H $^-$ ( $1^1S$ )	-0.527 735	0.527 751
Li $^+$ ( $1^1S$ )	-7.279 827	-7.279 913

TABLE I: Calculated energy levels in the helium atom and the helium-like ions H $^-$  and Li $^+$ , compared with reference values from Drake [2]. The calculations used  $l_{max} = 7$  and 50 exponentially distributed B-splines for each radial direction, in a box extending to 50 a.u.

### III. RESULTS

#### A. One photon double ionization

Because of the electron-electron interaction, double ionization of helium may proceed through the absorption of a single photon. This one-photon double ionization process has been investigated at length in both theoretical [2, 3, 8] and experimental [4, 44, 45] studies, resulting in close quantitative agreement in both total and differential ionization cross sections, and a thorough understanding of the physical underpinnings. This makes it an ideal benchmark against which new numerical schemes may be gauged. Accordingly, we have calculated cross sections for selected photon energies in the interval 80 – 200 eV. A box with  $r_{max} = 160$  a.u., 311 B-splines,  $l_{max} = 5$ , and values of  $L_{max}$  up to five was used. The pulse duration was set to 20 optical cycles, and the wave packet was propagated four additional cycles after the pulse was over, allowing the ionized component to reach the Coulomb zone. The results, shown in Fig. 1, include double ionization cross sections (red squares) and the ratio of double to single ionization (red circles), both within 5% of the experimental results of Samson *et al.* [4], who state the accuracy of their results to be  $\pm 2\%$ .

#### B. Two photon double ionization

We now consider the problem of two-photon direct double ionization. Our results, together with a small subset of results from the numerous studies available in the literature, are shown in Fig. 2. The calculations have been made using a box extending to  $r_{max} = 250$  a.u., and 246 B-splines, while the angular basis was truncated at  $l_{max} = 5$  and  $L_{max} = 3$ , respectively. Additionally, the intensity of the laser field was fixed at  $10^{13}$  W/cm $^2$ , which is well within the perturbative regime. We also checked that decreasing the intensity by a factor of ten did not produce any significant changes in the results; at  $\omega = 51.7$  eV the change in total cross section was less than 0.1%. Improving our basis by increasing  $(l_{1,max}, l_{2,max}, L_{max})$  to (7,7,5) and the number of B-splines to 270 resulted in only minor changes in the cross sections ( $< 0.03\%$  at 42 eV). To facilitate the separation of single-

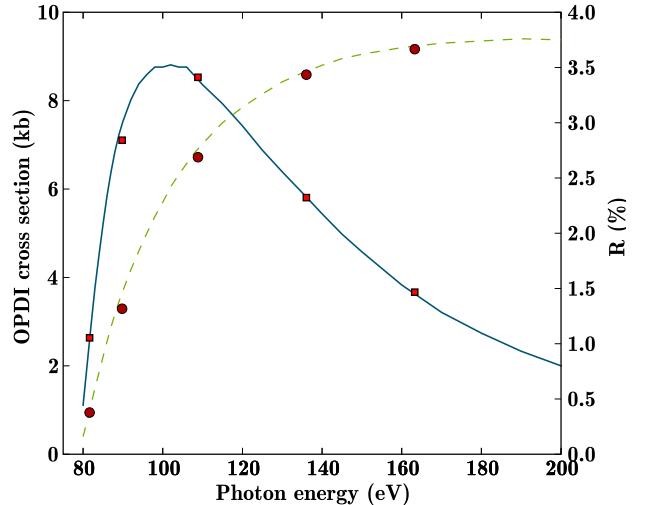


FIG. 1: (Color online) A comparison of calculated one-photon double ionization cross sections (red squares) and experimental values from Samson *et al.* [4] (blue full line). The ratio of double to single ionization is also shown. Red dots; calculated result, and green dashed line; experimental result.

and double continuum components of the wave function, we ran the propagation algorithm an additional femtosecond after the end of the pulse before performing the projections on the Coulomb waves, to ensure that the major part of the wave packet had entered the asymptotic Coulomb zone [18, 29].

The agreement between our results and those of Feist *et al.* [18] is particularly close, but this is not surprising due to the similarity of the numerical methods and the projection method used to extract the double ionization probability. In contrast, the J-matrix result of Foumouo *et al.* [5] (green line with diamonds in Fig. 2), and the perturbation theory result of Nikolopoulos *et al.* [17] (black line with triangles in Fig. 2), deviate significantly from ours, i.e., the calculated total cross sections for the reaction differ by as much as an order of magnitude. In both of these approaches, correlation effects were included in the final state, to some extent, while in our calculations no such effects were included. It should be noted, however, that Foumouo *et al.* [5] obtained similar results to ours when they neglected completely the role of electron-electron interactions in the final wave function (black line with crosses in Fig. 2). This seems to stress the importance of electron correlations in the final states, however, by propagating the wave packet for a long time after the pulse, the correlation effect should, in principle, be minimized, as argued and tested by Feist *et al.* [18]. In that particular study, they employed a very large grid and propagated the wave packet some 20 fs after the pulse, to explicitly check for convergence of the cross section. They also extracted the double ionization directly from the grid representation, by partitioning the radial grid, and by varying the partition limits found an upper bound for the possible value of the cross section  $\sim 25\%$  higher than their quoted results (Fig. 2).

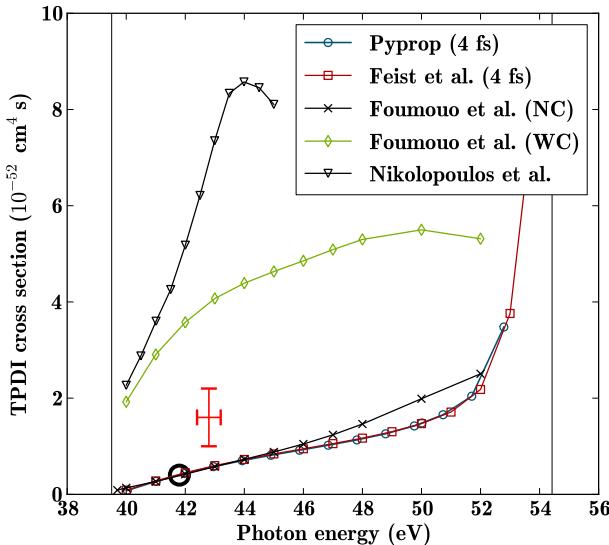


FIG. 2: (Color online) Two-photon double ionization cross sections. Blue line with circles: the present results obtained with Pyprop, black circle: experimental result of Hasegawa *et al.* [23], red cross: experimental result of Sorokin *et al.* [25], red line with squares: Feist *et al.* [18], green line with diamonds: Foumouo *et al.* [5] (with correlation, WC), black line with crosses: Foumouo *et al.* [5] (no correlation, NC), and black line with triangles: Nikolopoulos *et al.* [17]. The vertical lines define the two-photon direct double ionization region.

Regarding the apparent rapid rise in the value of the calculated cross section near the sequential ionization threshold (54.5 eV), this is usually attributed to the bandwidth of the pulses used in time-dependent methods and an unwanted inclusion of the sequential process [6, 16, 18, 21, 30]. Thus, due to the finite spectral width of the pulse, the sequential process cannot be completely separated from the nonsequential one, even below threshold. Now, since the relative importance of the sequential process scales as  $T^2$ , as opposed to the  $T$ -dependence of the nonsequential, attempting to extract a cross section when both processes are present would result in a divergent behavior. This problem can be circumvented by simply increasing the pulse duration in order to lower its bandwidth. Following this procedure, one avoids significant contribution from the sequential component up to some finite distance from the upper threshold, but at a certain point the overlap with the sequential region again becomes non-negligible, and the  $T^2$  scaling law causes an even sharper rise, due to the now longer pulse duration. Examining the relative importance of the different spectral components in the laser pulse as the pulse duration is increased, one can show that the relative contribution from the sequential process will ultimately become negligible, despite the  $T^2$ -dependence of the ionization yield. Thus, using successively longer pulses, one may, at least in principle, resolve the behavior of the direct two-photon double ionization cross section arbitrarily close to the threshold, without contamination from the sequential process.

Pursuing this line of thought, we have performed some ad-

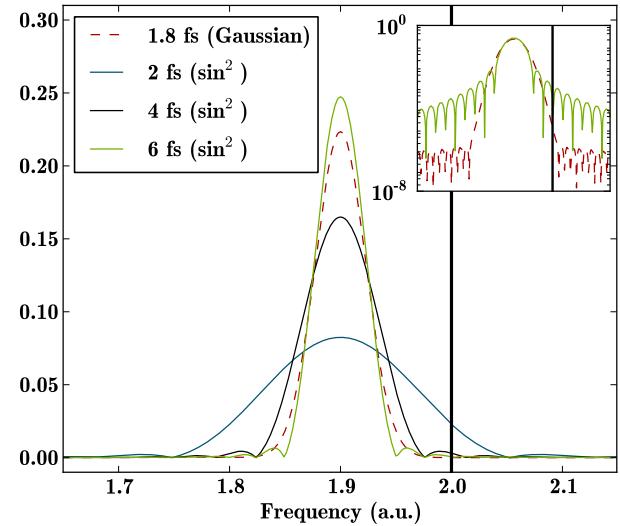


FIG. 3: (Color online) Fourier spectra of pulses with different temporal shapes and durations. Full lines represent sine-squared pulses with  $T = 2$  fs (blue), 4 fs (black) and 6 fs (green), while the red dashed line represents a Gaussian pulse with  $\tau_0 = 1.8$  fs and  $T = 9.4$  fs. The inset shows the 6 fs and Gaussian pulse spectra on a logarithmic scale.

Pulse duration (fs)	FWHM (fs)	Cross section ( $10^{-52} \text{ cm}^4 \text{s}$ )
2	0.7	2.6
4	1.5	2.1
5	1.8	2.0
6	2.2	2.0
9.4*	1.8	2.0

TABLE II: Double ionization cross sections at  $\hbar\omega = 51.7$  eV. (\*Gaussian pulse).

ditional calculations at  $\omega = 1.9$  a.u. (51.7 eV) with longer pulse durations and different temporal shapes, whose Fourier spectra are shown in Fig. 3. The 2 fs pulse has a clear extension into the sequential region, indicated by the black vertical line, while the longer pulses have successively less overlap. The resulting cross section values are shown in Table II. For pulse durations in the region 2 – 6 fs, and a sine-squared envelope, we found that the cross section leveled out at  $2.0 \times 10^{-52} \text{ cm}^4 \text{s}$  as  $T$  increased. Changing the temporal profile of the pulse to a Gaussian one, cf. Eq. 5, with a (total) duration of  $\sim 9.4$  fs, the same value for the cross section was obtained. In order to extract the cross section at photon energies exceeding 52 eV, significantly longer pulse durations than 5 fs would be required. Note that it was necessary to increase the radial box size to 350 a.u. (339 B-splines) in order to contain the double continuum wave packet when the pulse duration exceeded 4 fs.

#### IV. CONCLUSIONS

In this paper we have presented a numerical method for solving the two-electron time-dependent Schrödinger equation. After establishing the capability of the method through convergence tests and accurate reproduction of known physical quantities, we applied the method to the study of two-photon direct ionization of helium. Good agreement with several recently published results was found for the total cross section. Finally, we investigated the behavior of the cross section near the sequential threshold, where a steep increase was observed. Calculating the value of the cross section at fixed frequency (51.7 eV) for varying pulse duration, we found that the value converged for longer pulses, and it ap-

pears that the cross section indeed exhibits a moderate growing trend towards the threshold.

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